

What is Claimed is:

1. A method for the conversion of a carbon-based fuel to a H₂-rich product gas, comprising the steps of:

(a) providing a carbon-based fuel;

(b) converting said carbon-based fuel to an intermediate gas product by contacting said carbon-based fuel with at least a first conversion catalyst;

(c) contacting said intermediate gas product with an absorbent material to absorb CO₂ and form a H₂-rich gas, said absorbent material having a theoretical absorption capacity for CO₂;

(d) extracting said H₂-rich gas from said contacting step;

(e) regenerating said absorbent; and

(f) repeating said steps (a), (b), (c), (d) and (e) at least 10 times, wherein said absorbent material retains at least about 50 mol.% of said theoretical absorption capacity after each of said repeating steps.

2. A method as recited in Claim 1, wherein said converting step comprises steam reforming of said carbon-based fuel.

3. A method as recited in Claim 1, wherein said converting step is selected from the group consisting of autothermal reforming, partial oxidation and catalytic partial oxidation of said carbon-based fuel.

4. A method as recited in Claim 1, further comprising the step of contacting said H₂-rich gas with a water-gas shift catalyst.

5. A method as recited in Claim 1, wherein said repeating step comprises repeating steps (a), (b), (c), (d) and (e) at least 50 times.

6. A method as recited in Claim 1, wherein said repeating step comprises repeating steps (a), (b), (c), (d) and (e) at least 100 times.

7. A method as recited in Claim 1, wherein said repeating step comprises repeating steps (a), (b), (c), (d) and (e) at least 500 times.

8. A method as recited in Claim 1, wherein said absorbent material retains at least about 70 mol.% of said theoretical capacity after said repeating step.

9. A method as recited in Claim 1, wherein said absorbent material retains at least about 90 mol.% of said theoretical capacity after said repeating step.

10. A method as recited in Claim 1, wherein said repeating step comprises repeating steps (a), (b), (c), (d) and (e) at least 200 times and wherein said absorbent material retains at least about 10 mol.% of said theoretical absorption capacity after said repeating step.

5 11. A method as recited in Claim 1, wherein said repeating step comprises repeating steps (a), (b), (c), (d) and (e) at least 200 times and wherein said absorbent material retains at least about 25 mol.% of said theoretical absorption capacity after said repeating step.

10 12. A method as recited in Claim 1, wherein said repeating step comprises repeating steps (a), (b), (c) and (d) at least 200 times and wherein said absorbent material retains at least about 50 mol.% of said theoretical absorption capacity after said repeating step.

15 13. A method as recited in Claim 1, wherein said absorbent material comprises at least one metal oxide selected from the group consisting of Group IA and Group IIA metal oxides.

14. A method as recited in Claim 1, wherein said absorbent material comprises a calcium-containing compound.

15. A method as recited in Claim 1, wherein said absorbent material comprises CaO.

20 16. A method as recited in Claim 1, wherein said absorbent material comprises CaO:MgO.

17. A method as recited in Claim 1, wherein said absorbent material comprises CaO and from about 1 wt.% to about 40 wt.% Al_2O_3 .

25 18. A method as recited in Claim 1, wherein said contacting step occurs at a temperature of not greater than about 800°C.

19. A method as recited in Claim 1, wherein said carbon-based fuel is a hydrocarbon-based fuel.

20. A method as recited in Claim 1, wherein said carbon-based fuel is a gaseous fuel.

30 21. A method as recited in Claim 1, wherein said carbon-based fuel comprises methane.

22. A method as recited in Claim 1, wherein said carbon-based fuel comprises a liquid fuel.

23. A method as recited in Claim 1, wherein said carbon-based fuel comprises a liquid fuel selected from the group consisting of diesel fuel, JP-8 aviation fuel,
5 kerosene, ethanol and gasoline.

24. A method as recited in Claim 1, wherein said H₂-rich gas comprises at least about 95 mol.% H₂ after each said repeating steps.

25. A method as recited in Claim 1, wherein said regenerating step comprises heating said absorbent material to a temperature of at least about 700°C.

10 26. A method as recited in Claim 1, wherein said absorbent material is pelletized.

27. A method as recited in Claim 1, wherein said absorbent material is coated on a support structure.

28. A method as recited in Claim 1, wherein said absorbent material has
15 substantially spherical morphology

29. A method for the conversion of a carbon-based fuel to a H₂-rich product gas, comprising the steps of:

(a) providing a carbon-based fuel;

(b) converting said carbon-based fuel to an intermediate gas product by contacting said carbon-based fuel with at least a first conversion catalyst;

(c) contacting said intermediate gas product with an absorbent material to absorb CO₂ and form a H₂-rich gas, said absorbent material having an theoretical absorption capacity for CO₂;

(d) extracting said H₂-rich gas from said contacting step;

(e) regenerating said absorbent; and

(f) repeating said steps (a), (b), (c), (d) and (e) at least 10 times, wherein said total mass of absorbent material retains at least about 10 grams CO₂ per 100 grams unreacted absorbent after each of said repeating steps.

30. A method as recited in Claim 29, wherein said absorbent material retains at least 20 grams CO₂ per 100 grams unreacted absorbent after each of said repeating steps.

31. A method as recited in Claim 29, wherein said absorbent material retains at least 30 grams CO₂ per 100 grams unreacted absorbent after each of said repeating steps.

32. A method as recited in Claim 29, wherein said absorbent material retains at least 40 grams CO₂ per 100 grams unreacted absorbent after each of said repeating steps.

33. A method as recited in Claim 29, wherein said absorbent material retains at least 50 grams CO₂ per 100 grams unreacted absorbent after each of said repeating steps.

34. A method as recited in Claim 29, wherein said absorbent material is pelletized.

35. A method as recited in Claim 29, wherein said converting step comprises steam reforming of said carbon-based fuel.

36. A method as recited in Claim 29, wherein said converting step is selected from the group consisting of autothermal reforming, partial oxidation and catalytic partial oxidation of said carbon-based fuel.

5 37. A method as recited in Claim 29, further comprising the step of contacting said H₂-rich gas with a water-gas shift catalyst.

38. A method as recited in Claim 29, wherein said repeating step comprises repeating steps (a), (b), (c), (d) and (e) at least 50 times.

39. A method as recited in Claim 29, wherein said repeating step comprises repeating steps (a), (b), (c), (d) and (e) at least 100 times.

10 40. A method as recited in Claim 29, wherein said repeating step comprises repeating steps (a), (b), (c), (d) and (e) at least 500 times.

41. A method as recited in Claim 29, wherein said absorbent retains at least about 70 mol.% of said theoretical absorption capacity after said repeating step.

15 42. A method as recited in Claim 29, wherein said absorbent retains at least about 90 mol.% of said theoretical absorption capacity after said repeating step.

43. A method as recited in Claim 29, wherein said repeating step comprises repeating steps (a), (b), (c), (d) and (e) at least 200 times and wherein said absorbent material retains at least about 10 mol.% of said theoretical absorption capacity after said repeating step.

20 44. A method as recited in Claim 29, wherein said repeating step comprises repeating steps (a), (b), (c), (d) and (e) at least 200 times and wherein said absorbent material retains at least about 25 mol.% of said theoretical absorption capacity after said repeating step.

25 45. A method as recited in Claim 29, wherein said repeating step comprises repeating steps (a), (b), (c) and (d) at least 200 times and wherein said absorbent material retains at least about 50 mol.% of said theoretical absorption capacity after said repeating step.

30 46. A method as recited in Claim 29, wherein said absorbent material comprises at least one metal-oxide selected from the group consisting of Group IA and Group IIA metal oxides.

47. A method as recited in Claim 29, wherein said absorbent material comprises a calcium-containing compound.

48. A method as recited in Claim 29, wherein said absorbent material comprises CaO.

5 49. A method as recited in Claim 29, wherein said absorbent material comprises CaO:MgO.

50. A method as recited in Claim 29, wherein said absorbent material comprises CaO and from about 1 wt.% to about 40 wt.% Al₂O₃.

10 51. A method as recited in Claim 29, wherein said contacting step occurs at a temperature of not greater than about 800°C.

52. A method as recited in Claim 29, wherein said carbon-based fuel is a hydrocarbon-based fuel.

53. A method as recited in Claim 29, wherein said carbon-based fuel is a gaseous fuel.

15 54. A method as recited in Claim 29, wherein said carbon-based fuel comprises methane.

55. A method as recited in Claim 29, wherein said carbon-based fuel is a liquid fuel.

20 56. A method as recited in Claim 29, wherein said carbon-based fuel comprises a liquid selected from the group consisting of diesel fuel, JP-8 aviation fuel, kerosene, ethanol and gasoline.

57. A method as recited in Claim 29, wherein said H₂-rich gas comprises at least about 95 mol.% H₂ after said repeating step.

25 58. A method as recited in Claim 29, wherein said regenerating step comprises heating said absorbent material to a temperature of at least about 700°C.

59. A method as recited in Claim 29, wherein said absorbent material is pelletized.

60. A method as recited in Claim 29, wherein said absorbent material is coated on a support structure.

30 61. A method as recited in Claim 29, wherein said absorbent material has substantially spherical morphology.

62. A method for the conversion of a carbon-based fuel to a H₂-rich gas, comprising the steps of:

(a) providing a carbon-based fuel and steam;

(b) converting said carbon-based fuel and said steam to an intermediate gas product by contacting with at least a first conversion catalyst;

(c) contacting said intermediate gas product with an absorbent material to absorb CO₂ and form an H₂-rich gas, said absorbent material having a theoretical absorption capacity and wherein at least said absorbent material is pelletized;

(d) extracting said H₂-rich gas from said contacting step;

(e) regenerating said absorbent; and

(f) repeating said steps (a), (b), (c), (d) and (e) at least 50 times, wherein said absorbent material retains at least about 20 mol.% of its theoretical CO₂ absorption capacity after each of said repeating steps.

63. A method as recited in Claim 62, wherein said absorbent material retains at least about 40 mol.% of said theoretical CO₂ absorption capacity after each of said repeating steps.

64. A method as recited in Claim 62, wherein said absorbent material retains at least about 60 mol.% of said theoretical CO₂ absorption capacity after each of said repeating steps.

65. A method as recited in Claim 62, wherein said absorbent material retains at least about 90 mol.% of said theoretical CO₂ absorption capacity after each of said repeating steps.

66. A method as recited in Claim 62, wherein said absorbent material retains at least 10 grams CO₂ per 100 grams unreacted absorbent material after each of said repeating steps.

67. A method as recited in Claim 62, wherein said absorbent material retains at least 20 grams CO₂ per 100 grams unreacted absorbent material after each of said repeating steps.

68. A method as recited in Claim 62, wherein said absorbent material retains at least 30 grams CO₂ per 100 grams unreacted absorbent material after each of said repeating steps.

5 69. A method as recited in Claim 62, wherein said absorbent material retains at least 40 grams CO₂ per 100 grams unreacted absorbent material after each of said repeating steps.

70. A method as recited in Claim 62, wherein said absorbent material is regenerated at least about 100 times and wherein said absorbent material retains at least about 20 mol.% of said theoretical CO₂ absorption capacity.

10 71. A method as recited in Claim 62, wherein said absorbent material has substantially spherical morphology

72. A method as recited in Claim 62, wherein said first conversion catalyst is a steam reforming catalyst.

15 73. A method as recited in Claim 62, further comprising the step of contacting said H₂-rich product gas with a water-gas shift catalyst.

74. A method as recited in Claim 62, wherein said absorbent material comprises at least one metal oxide selected from the group consisting of Group IA and Group IIA metal oxides.

20 75. A method as recited in Claim 62, wherein said absorbent material comprises a calcium-containing compound.

76. A method as recited in Claim 62, wherein said absorbent material comprises CaO.

77. A method as recited in Claim 62, wherein said absorbent material comprises CaO and from about 1 wt.% to about 40 wt.% Al₂O₃.

25 78. A method as recited in Claim 62, wherein said absorbent comprises CaO and a metal oxide selected from the group consisting of Al₂O₃ and MgO.

79. A method as recited in Claim 62, wherein said first conversion catalyst comprises a metal selected from the group consisting of Rh, Ni, Ru, Pt, and Pd.

30 80. A method as recited in Claim 62, wherein said contacting step occurs at a temperature of not greater than about 800°C.

81. A method as recited in Claim 62, wherein said regenerating step comprises heating said absorbent material to a temperature of at least about 700°C.

82. A method as recited in Claim 62, wherein said conversion catalyst is pelletized with said absorbent material.

5 83. A method as recited in Claim 62, wherein said H₂-rich product gas comprises at least about 95 mol.% H₂.

84. A method for absorption enhanced reforming of a carbon-based fuel, comprising the steps of:

(a) providing a carbon-based fuel;

(b) contacting said carbon-based fuel with at least a first conversion catalyst to catalyze the formation of an intermediate gas product;

(c) contacting said intermediate gas product with a pelletized absorbent compound having a first bulk density, wherein said pelletized absorbent is converted to a carbonized absorbent having a second bulk density; and

(d) regenerating said carbonized absorbent to form a regenerated absorbent having a third bulk density, wherein said third bulk density is greater than said first bulk density.

85. A method as recited in Claim 84, wherein said third bulk density is up to about 140% of said first bulk density.

86. A method as recited in Claim 84, wherein said regenerating step comprises heating said carbonized absorbent to a temperature of at least about 700°C.

87. A method as recited in Claim 84, wherein said carbon-based fuel is a hydrocarbon-based fuel.

88. A method as recited in Claim 84, wherein said carbon-based fuel is gaseous fuel.

89. A method as recited in Claim 84, wherein said carbon-based fuel comprises methane.

90. A method as recited in Claim 84, wherein said carbon-based fuel is a liquid fuel.

91. A method as recited in Claim 84, wherein said carbon-based fuel is selected from the group consisting of gasoline, kerosene, jet fuel, bio-oil from carbon.

92. A method as recited in Claim 84, further comprising the step of providing steam with said carbon-based fuel.

93. A method as recited in Claim 84, further comprising the step of providing steam, or an oxygen-containing gas with said carbon-based fuel.

94. A method as recited in Claim 84, wherein said absorbent compound comprises a calcium-containing compound.

95. A method as recited in Claim 84, wherein said absorbent compound comprises CaO.

5 96. A method as recited in Claim 84, wherein said absorbent compound comprises CaO:MgO.

97. A method as recited in Claim 84, wherein said absorbent has substantially spherical morphology.

10 98. A method as recited in Claim 84, wherein said regenerating step comprises heating said absorbent compound to a temperature of at least about 700°C.

99. A steam-reforming catalyst, comprising:

(a) a particulate support structure; and

(b) a metal dispersed on said support structure, wherein said reforming catalyst achieves at least about 90% of the theoretical thermodynamic conversion of methane to hydrogen at a temperature of 600°C, a H₂O:C ratio of 3:1 and a gas hour space velocity (GHSV) of 5000 hour⁻¹ in the absence of an absorbent for CO₂.

100. A reforming catalyst as recited in Claim 99, wherein said catalyst achieves at least about 95% of the theoretical thermodynamic conversion.

101. A reforming catalyst as recited in Claim 99, wherein said catalyst achieves at least about 90% of the theoretical thermodynamic conversion of methane to hydrogen at a temperature of 600°C, a H₂O:C ratio of 3:1 and a gas hour space velocity (GHSV) of 10000 hour⁻¹ in the absence of an absorbent for CO₂.

102. A reforming catalyst as recited in Claim 99, wherein said catalyst achieves at least about 90% of the theoretical thermodynamic conversion of methane to hydrogen at a temperature of 600°C, a H₂O:C ratio of 3:1 and a gas hour space velocity (GHSV) of 12500 hour⁻¹ in the absence of an absorbent for CO₂.

103. A reforming catalyst as recited in Claim 99, wherein said support is selected from the group consisting of the metal oxides of aluminum, cerium, zirconium, lanthanum, silicon, magnesium, zinc and combinations thereof.

104. A reforming catalyst as recited in Claim 99, wherein said dispersed metal is selected from the group consisting of Rh, Ni, Ru, Pt, Pd and alloys thereof.

105. A reforming catalyst as recited in Claim 99, wherein said dispersed metal comprises Rh.

106. A reforming catalyst as recited in Claim 99, wherein said reforming catalyst comprises from about 0.1 wt.% to about 1 wt.% of said metal.

107. A reforming catalyst as recited in Claim 99, wherein said support structure comprises Al₂O₃ and said dispersed metal comprises Rh.

108. A reforming catalyst as recited in Claim 99, wherein said reforming catalyst is pelletized.

109. A reforming catalyst as recited in Claim 99, wherein said reforming catalyst is coated on a support.

110. A reforming catalyst as recited in Claim 99, wherein said reforming catalyst has substantially spherical morphology.

111. A particulate composite material, said particulates comprising:

(a) an absorbent phase adapted to absorb CO₂; and

(b) a catalyst phase selected from the group consisting of a reforming catalyst phase and a water-gas shift catalyst phase.

5 112. A particulate composite material as recited in Claim 111, wherein said absorbent phase comprises a calcium compound.

113. A particulate composite material as recited in Claim 111, wherein the mass ratio of said absorbent phase to said catalyst phase is greater than 1:1.

10 114. A particulate composite material as recited in Claim 111, wherein the mass ratio of said absorbent phase to said catalyst phase is from about 20:1 to about 3:1.

115. A particulate composite material as recited in Claim 111, wherein the mass ratio of said absorbent phase to said catalyst phase is from about 9:1 to about 5:1.

15 116. A particulate composite as recited in Claim 111, wherein said catalyst phase comprises a supported metal.

117. A particulate composite material as recited in Claim 111, wherein said catalyst phase comprises a reforming catalyst.

20 118. A particulate composite material as recited in Claim 111, wherein said catalyst is a reforming catalyst comprising a metal selected from Rh, Ni, Ru, Pt, Pd, and alloys thereof dispersed on a support phase selected from the group consisting of metal oxides of aluminum, cerium, zirconium, lanthanum, silicon, magnesium, zinc and combinations thereof.

25 119. A particulate composite material as recited in Claim 111, wherein said catalyst phase comprises a water-gas shift catalyst.

120. A particulate composite material as recited in Claim 111, wherein said catalyst is a water-gas shift catalyst comprising a metal dispersed on a support phase, said metal being selected from the group consisting of Fe, Co, Cu and Cr.

30 121. A particulate composite material as recited in Claim 111, wherein said particulate composite material is pelletized.

122. A particulate composite material as recited in Claim 111, wherein said particulate composite material is coated on a support structure.

123. A particulate composite material as recited in Claim 111, wherein said absorbent phase includes an absorbent compound having a reaction fraction of at least about 70 mol.%.
5

124. A particulate composite material as recited in Claim 111, wherein said absorbent material has a reaction fraction of at least about 70 mol.% after 100 cycles.

125. A particulate composite material as recited in Claim 111, wherein said absorbent phase comprises a reversible absorbent.

126. A particulate composite material as recited in Claim 111, wherein said absorbent phase comprises an absorbent compound that retains at least about 40 mol.% of the theoretical CO₂ absorption capacity of the absorbent compound after at least about 10 cycles.
10

127. A particulate composite material as recited in Claim 111, wherein the said absorbent phase comprises an absorbent compound that retains at least about 60 mol.% of the theoretical CO₂ absorption capacity of the absorbent compound after at least about 10 cycles.
15

128. A particulate composite material as recited in Claim 111, wherein said absorbent phase comprises an absorbent compound retains at least about 90 mol.% of the theoretical CO₂ absorption capacity of the absorbent compound after at least about 10 cycles.
20

129. A particulate composite material as recited in Claim 111, wherein said absorbent phase retains at least 10 grams CO₂ per 100 grams unreacted absorbent after 10 cycles.

130. A particulate composite material as recited in Claim 111, wherein said absorbent retains at least 20 grams CO₂ per 100 grams unreacted absorbent after 10 cycles.
25

131. A particulate composite material as recited in Claim 111, wherein said absorbent retains at least 30 grams CO₂ per 100 grams unreacted absorbent after 10 cycles.
30

132. A particulate composite material as recited in Claim 111, wherein said absorbent retains at least 40 grams CO₂ per 100 grams unreacted absorbent after 10 cycles.

5 133. A particulate composite material as recited in Claim 111, wherein said absorbent retains at least 50 grams CO₂ per 100 grams unreacted absorbent after 10 cycles.

134. A particulate composite material as recited in Claim 111, wherein said particulate composite has substantially spherical morphology.

135. A method for the fabrication of supported reforming catalyst particles, comprising the steps of:

(a) forming a precursor solution comprising a metal precursor and an alumina precursor;

(b) atomizing said precursor solution to form precursor droplets;

(c) heating said precursor droplets to convert at least said metal precursor to metal-containing clusters dispersed on said alumina.

136. A method as recited in Claim 135, wherein said metal precursor comprises a precursor to a metal selected from the group consisting of Rh, Ni, Ru, Pt, Pd and alloys thereof.

137. A method as recited in Claim 135, wherein said metal precursor comprises a precursor to Rh.

138. A method as recited in Claim 135, wherein said alumina precursor comprises particulate alumina.

139. A method as recited in Claim 135, wherein said alumina precursor comprises particulate boehmite alumina.

140. A method as recited in Claim 135, wherein said alumina precursor comprises particulate alumina having an average particle size of not greater than about 100 nm.

141. A method as recited in Claim 135, wherein said catalyst particles have an average particle size (d_{50}) of from about $1\mu\text{m}$ to about $30\mu\text{m}$.

142. A method as recited in Claim 135, wherein said catalyst particles have a surface area of at least about $150\text{ m}^2/\text{g}$.

143. A method as recited in Claim 135, wherein said catalyst particles are aggregate particles comprising aggregated primary support particles, said primary support particles having an average size of from about 10 nanometers to about 100 nanometers.

144. A method as recited in Claim 135, wherein said heating step comprises heating said precursor droplets to a temperature of at least about 500°C .

145. A method as recited in Claim 135, wherein said catalyst particles comprise from about 0.1 wt.% to about 5 wt.% of said metal-containing clusters.

146. A method as recited in Claim 135, wherein said heating step is carried out in a spray dryer.

5 147. A method as recited in Claim 135, further comprising the step of heating said particles to a temperature of at least about 500°C.

148. A method as recited in Claim 135, wherein said reforming particles have substantially spherical morphology.

149. A method for the fabrication of composite particles including an absorbent phase and a catalyst phase comprising the steps of:

(a) forming a precursor solution, said precursor solution comprising:

(i) a liquid;

(ii) a precursor to an absorbent phase; and

(iii) a precursor to a catalyst phase;

(b) atomizing said precursor solution to form precursor droplets; and

(c) heating said precursor droplets to remove said liquid and form composite particles.

150. A method as recited in Claim 149, wherein said heating step comprises a first heating step to react said active absorbent precursor to an intermediate precursor compound and a second heating step to convert said intermediate precursor compound to said absorbent material.

151. A method as recited in Claim 150, wherein said second heating step is at a temperature that is higher than said first heating step.

152. A method as recited in Claim 149, wherein said heating step comprises heating said precursor mixture to form said absorbent material in a single step.

153. A method as recited in Claim 149, wherein said absorbent phase precursor is selected from the group consisting of calcium oxalate, calcium nitrate, calcium acetate, calcium lactate and calcium hydroxide.

154. A method as recited in Claim 149, wherein said absorbent phase precursor comprises calcium nitrate.

155. A method as recited in Claim 149, wherein said precursor solution further comprises a porosity enhancing agent.

156. A method as recited in Claim 149, wherein said catalyst phase precursor comprises a precursor to a metal selected from the group consisting of Rh, Ni, Ru, Pt, Pd and alloys thereof.

157. A method as recited in Claim 149, wherein said catalyst phase precursor comprises a precursor to Rh.

158. A method as recited in Claim 149, wherein said catalyst phase precursor comprises particulate alumina.

159. A method as recited in Claim 149, wherein said composite particles have an average particle size (d_{50}) of from about 1 μm to about 30 μm .

160. A method as recited in Claim 149, wherein said heating step comprises heating said precursor droplets to a temperature of at least about 200°C.

5 161. A method as recited in Claim 149, wherein said heating step is carried out in a spray dryer.

162. A method as recited in Claim 149, wherein said composite particles have substantially spherical morphology.

163. A method for the fabrication of composite particles including an absorbent phase and a catalyst phase, comprising the steps of:

(a) forming a particulate suspension, said particulate suspension comprising:

(i) a liquid;

(ii) a first particulate phase comprising an absorbent phase or an intermediate precursor to an absorbent phase; and

(iii) a second particulate phase comprising a catalyst phase;

(b) atomizing said particulate suspension to form suspension droplets;

and

(c) heating said suspension droplets to remove said liquid and form composite particles.

164. A method as recited in Claim 163, wherein said heating step comprises a first heating step to react said active absorbent precursor to an intermediate precursor compound and a second heating step to convert said intermediate precursor compound to said absorbent material.

165. A method as recited in Claim 163, wherein said second heating step is at a temperature that is higher than said first heating step.

166. A method as recited in Claim 163, wherein said heating step comprises heating said precursor mixture to form said absorbent material in a single step.

167. A method as recited in Claim 163, wherein said absorbent phase precursor is selected from the group consisting of calcium oxalate, calcium nitrate, calcium acetate, calcium lactate and calcium hydroxide.

168. A method as recited in Claim 163, wherein said absorbent phase precursor comprises calcium nitrate.

169. A method as recited in Claim 163, wherein said precursor solution further comprises a porosity enhancing agent.

170. A method as recited in Claim 163, wherein said catalyst phase precursor comprises a precursor to a metal selected from the group consisting of Rh, Ni, Ru, Pt, Pd and alloys thereof.

171. A method as recited in Claim 163, wherein said catalyst phase precursor comprises a precursor to Rh.

172. A method as recited in Claim 163, wherein said catalyst phase precursor comprises particulate alumina.

5 173. A method as recited in Claim 163, wherein said composite particles have an average particle size (d_{50}) of from about 1 μm to about 30 μm .

174. A method as recited in Claim 163, wherein said heating step comprises heating said precursor droplets to a temperature of at least about 200°C.

10 175. A method as recited in Claim 163, wherein said heating step is carried out in a spray dryer.

176. A method as recited in Claim 163, wherein said composite particles have substantially spherical morphology.

177. A method as recited in Claim 163, wherein said first particulate phase has substantially spherical morphology.

15 178. A method as recited in Claim 163, wherein said second particulate phase has substantially spherical morphology.

179. A pellet adapted for absorption enhanced reforming of a carbon-based fuel, said pellet comprising:

- (a) a particulate absorbent material; and
- (b) a particulate catalyst material.

5 180. A pellet as recited in Claim 179, wherein said absorbent material comprises a calcium-containing compound.

181. A pellet as recited in Claim 179, wherein said absorbent comprises CaO.

182. A pellet as recited in Claim 179, wherein said catalyst is a reforming catalyst comprising a metal selected from the group consisting of Rh, Ni, Ru, Pt, Pd and
10 alloys thereof dispersed on a support phase.

183. A pellet as recited in Claim 179, wherein the mass ratio of said absorbent material to said catalyst material is at least about 1:1.

184. A pellet as recited in Claim 179, wherein the mass ratio of said absorbent material to said catalyst material is from about 20:1 to about 3:1.

15 185. A pellet as recited in Claim 179, wherein the mass ratio of said absorbent material to said catalyst material is from about 9:1 to about 5:1.

186. A pellet as recited in Claim 179, wherein said pellet further comprises a binder.

187. A pellet as recited in Claim 186, wherein said binder comprises aluminum
20 oxide.

188. A pellet as recited in Claim 179, wherein said particulate absorbent material comprises an active absorbent phase and an inert metal oxide phase.

189. A pellet as recited in Claim 179, wherein said particulate absorbent material has substantially spherical morphology.

25 190. A pellet as recited in Claim 179, wherein said particulate catalyst material has substantially spherical morphology.

191. A pellet adapted for absorption enhanced reforming of a carbon-based fuel, said pellet comprising composite particulates including an absorbent phase adapted to absorb CO₂ and a catalyst phase selected from the group consisting of a reforming catalyst and a water-gas shift catalyst.

5 192. A pellet as recited in Claim 191, wherein said absorbent phase comprises a calcium-containing compound.

193. A pellet as recited in Claim 191, wherein said absorbent phase comprises CaO.

10 194. A pellet as recited in Claim 191, wherein said catalyst phase is a steam-methane reforming catalyst.

195. A pellet as recited in Claim 191, wherein said catalyst phase is a reforming catalyst comprising a metal selected from the group consisting of Rh, Ni, Ru, Pt, Pd and alloys thereof dispersed on a support phase.

15 196. A pellet as recited in Claim 191, wherein the mass ratio of said absorbent phase to said catalyst phase is at least about 1:1.

197. A pellet as recited in Claim 191, wherein the mass ratio of said absorbent phase to said catalyst phase is from about 20:1 to about 3:1.

198. A pellet as recited in Claim 191, wherein the mass ratio of said absorbent phase to said catalyst phase is from about 9:1 to about 5:1.

20 199. A pellet as recited in Claim 191, wherein said pellet has a crush strength of at least about 1 N/mm.

200. A pellet as recited in Claim 191, wherein said pellet further comprises a binder.

25 201. A pellet as recited in Claim 191, wherein said absorbent phase has substantially spherical morphology.

202. A pellet as recited in Claim 191, wherein said catalyst phase has substantially spherical morphology.